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(11) **EP 0 748 824 A1**

(12) **EUROPEAN PATENT APPLICATION**

(43) Date of publication:
18.12.1996 Bulletin 1996/51

(51) Int. Cl.⁶: **C08F 10/06**

(21) Application number: 96108226.0

(22) Date of filing: 23.05.1996

(84) Designated Contracting States:
BE DE FR GB IT NL

(30) Priority: 02.06.1995 JP 136425/95

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(54) **High stereoregular polypropylenes**

(57) A high stereoregular polypropylene characterized in that

- (a) an isotactic pentad (m m m m) is 0.950 to 0.995,
- (b) a syndiotactic pentad (r r r r) is 0 to 0.01,
- (c) a different bond due to 2,1- and 1,3-insertions of propylene monomer is 0 to 0.3 mol%,
- (d) an absence of a terminal double bond is confirmed,
- (e) a weight average molecular weight (Mw) is 50,000 to 1,000,000 and
- (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is 1.5 to 3.8, (a) to (d) being determined from ¹³C NMR spectra.

The high stereoregular polypropylenes have narrow molecular weight distributions and high melting points, which exhibit very high tenacity, high-temperature rigidity, heat resistance and good molding properties when used as molding materials.

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Description

FIELD OF THE INVENTION

5 This invention relates to high stereoregular polypropylenes. More specifically, the invention relates to the high stereoregular polypropylenes with narrow molecular weight distributions and high melting points, which exhibit very high tenacity, high-temperature rigidity, heat resistance and good molding properties when used as molding materials, to processes for the production of such polypropylenes and also molded products formed from such polypropylenes.

10 BACKGROUND OF THE INVENTION

Crystalline polypropylenes have been extensively used in the field of various moldings, because of good mechanical properties, good chemical resistance or the like, very useful balance with economy. However, conventional known polypropylenes have limited use. Thus a strong desire has been directed to the improvement in performance of polypropylenes, especially the improvement in heat resistance characteristics such as high-temperature rigidity and high tenacity.

15 In recent years, it is known that propylene is polymerized using a catalyst consisting of combinations of metallocenes with aluminoxanes to produce stereoregular polypropylenes with narrow molecular weight distributions. For instance, propylene is subjected to polymerization using a catalyst consisting of silylene-bridged metallocenes and aluminoxanes having the specific structures to produce high stereoregular polypropylenes with narrow molecular weight distributions, as disclosed in Japanese Patent Kokai 3-12406 and 3-12407 and CHEMISTRY LETTERS, PP. 1853-1856, 1989.

20 The polypropylenes produced by the above processes have narrow molecular weight distribution and high stereoregularity, and also higher melting point and higher rigidity than polypropylenes produced from the use of prior metallocene catalysts, but they have a double bond at one end of the polymer and may impair a chemical stability depending on the use condition, since the polymerization of propylene is performed in the absence of hydrogen. Thus the improvement of such processes has been desired with more improved heat resistance and higher tenacity.

25 Polypropylenes with narrow molecular weight distributions produced by known processes, due to the presence of a double bond at one end, have the problem that they are required to improve a chemical stability and a heat resistance and provide a high tenacity.

SUMMARY OF THE INVENTION

35 The present inventors have made extensive investigation in an effort to solve the above-mentioned problem and as a result have succeeded in producing polypropylenes with narrow molecular weight distribution and high stereoregularity having no double bond, substantially no different bond, and also have found that polypropylenes with such particular structure have exceedingly high high-temperature characteristics, high tenacity and also good molding property. Further, they have found that polypropylenes having such structure can be produced only under the specified polymerization conditions, regardless of using similar catalysts to those used in the above prior art. Such findings lead to the completion of the invention.

40 As apparent from the foregoing, an object of the present invention is to solve the above-mentioned problems and to provide high stereoregular polypropylenes with narrow molecular weight distributions and high melting points, which exhibit very high tenacity, high-temperature rigidity, heat resistance and good molding properties when used as molding materials.

45 According to the present invention, there is provided a high stereoregular polypropylene characterized in that

- (a) an isotactic pentad (m m m m) is 0.950 to 0.995,
- (b) a syndiotactic pentad (r r r r) is 0 to 0.01,
- (c) a different bond due to 2,1- and 1,3-insertions of propylene monomer is 0 to 0.3 mol%,
- 50 (d) an absence of a terminal double bond is confirmed,
- (e) a weight average molecular weight (Mw) is 50,000 to 1,000,000 and
- (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is 1.5 to 3.8, (a) to (d) being determined from ¹³C NMR spectra.

55 In another aspect of the present invention, there is provided a process for the production of a high stereoregular polypropylene characterized in that

- (a) an isotactic pentad (m m m m) is 0.950 to 0.995,
- (b) a syndiotactic pentad (r r r r) is 0 to 0.01,

- (c) a different bond due to 2,1- and 1,3-insertions of propylene monomer is 0 to 0.3 mol%,
 (d) an absence of a terminal double bond is confirmed,
 (e) a weight average molecular weight (Mw) is 50,000 to 1,000,000 and
 (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is 1.5 to 3.8, (a) to (d) being determined from ^{13}C NMR spectra, wherein propylene is polymerized in the presence of hydrogen using a catalyst comprising a chiral transition metal compound and an aluminosilane, the transition metal compound being represented by the formula of $\text{Q}(\text{C}_5\text{H}_4\text{-mR}^1\text{m})(\text{C}_5\text{H}_4\text{-nR}^2\text{n})\text{MXY}$ wherein $(\text{C}_5\text{H}_4\text{-mR}^1\text{m})$ and $(\text{C}_5\text{H}_4\text{-nR}^2\text{n})$ represent a substituted cyclopentadienyl group; m and n are an integer of 1 to 3; R^1 and R^2 may be the same or different and each represents a hydrocarbyl radical of 1 to 20 carbons, a silicone-containing hydrocarbyl radical or a hydrocarbyl radical which is joined with two carbon atoms on the cyclopentadienyl ring to form one or more hydrocarbon rings which may be substituted by a hydrocarbon; Q is a group capable of crosslinking $(\text{C}_5\text{H}_4\text{-mR}^1\text{m})$ and $(\text{C}_5\text{H}_4\text{-nR}^2\text{n})$ and represents a divalent, hydrocarbyl radical, an unsubstituted silylene group or a hydrocarbyl-substituted silylene group; M represents a transition metal selected from the group consisting of titanium, zirconium and hafnium; X and Y may be the same or different and each represents a hydrogen, a halogen or a hydrocarbyl radical.

In other aspects of the present invention, there is provided a molded product formed from a high stereoregular polypropylene characterized in that

- (a) an isotactic pentad (m m m m) is 0.950 to 0.995,
 (b) a syndiotactic pentad (r r r r) is 0 to 0.01,
 (c) a different bond due to 2,1- and 1,3-insertions of propylene monomer is 0 to 0.3 mol%,
 (d) an absence of a terminal double bond is confirmed,
 (e) a weight average molecular weight (Mw) is 50,000 to 1,000,000 and
 (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is 1.5 to 3.8, (a) to (d) being determined from ^{13}C NMR spectra.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 shows ^{13}C NMR spectra of the high stereoregular polypropylene of the present invention produced in Example 1.

DETAILED DESCRIPTION OF THE INVENTION

Of the requirements for characterizing the stereoregular polypropylenes of the present invention, the above-identified (a), (b), (c) and (d) are calculated based on the results determined by ^{13}C NMR spectra in accordance with the following method. More specifically, the NMR spectra was recorded at 130°C on a JEOL-GX270 spectrometer manufactured by Nihon Densi K.K. in Japan operating at 67.20 MHz, using a mixed solution of o-dichlorobenzene/benzene bromide with 8/2 weight ratio having 20% by weight of polymer concentration.

The terms "isotactic pentad (m m m m)" and "syndiotactic pentad (r r r r)" as used herein refer to the isotactic sequence and the syndiotactic sequence, respectively, in terms of a pentad unit in the polypropylene molecular chain determined by ^{13}C NMR spectra which was suggested by A. Zambelli, et al in *Macromolecules* Vol. 6, No. 6, 925-926, 1973.

A method of deciding the assignment of peak in the determination of ^{13}C NMR spectra is performed in accordance with the assignment suggested by A. Zambelli, et al in *Macromolecules* Vol. 8, No. 5, 687-688, 1975.

The isotactic pentad (m m m m) (a) of the requirements for characterizing the stereoregular polypropylene of the present invention is the proportion of a propylene monomer unit containing 5 successive meso bonds which are present in all propylene monomer units in the polypropylene molecules. Higher isotactic (m m m m) pentad shows higher isotactic characteristics. For the polypropylenes of the present invention, the isotactic (m m m m) pentad is 0.950 to 0.995, preferably 0.955 to 0.995, especially preferably 0.960 to 0.995.

The syndiotactic pentad (r r r r) (b) of the requirements for characterizing the stereoregular polypropylene of the present invention is the proportion of a propylene monomer unit containing 5 successive racemic bonds which are present in all propylene monomer units in the polypropylene molecules. Lower syndiotactic (r r r r) pentad shows lower syndiotactic characteristics. For the polypropylenes of the present invention, the syndiotactic (r r r r) pentad is 0 to 0.01, preferably 0 to 0.007, especially preferably 0 to 0.004.

The term "different bond due to 2,1- and 1,3-insertions of propylene monomer" as used herein refers to the proportion of a different bond due to 2,1- and 1,3-insertions of propylene monomer which is present in the polypropylene molecules determined by ^{13}C NMR spectra in accordance with the method suggested by T. Tsutsui et al in *POLYMER* Vol. 30, 1350-1356, 1989.

The different bond due to 2,1- and 1,3-insertions of propylene monomer (c) of the requirements for characterizing the stereoregular polypropylene of the present invention is 0 to 0.3 mol%, preferably 0 to 0.25 mol%, especially preferably 0 to 0.2 mol%. When conventional known titanium catalysts are used in the polymerization of propylene, the polymerization proceeds by the 2,1-insertion. When known metallocene catalysts are used, on the other hand, it is known that a certain degree of 2,1- and 1,3-insertions takes place and a specific amount of different bond is present in the resulting polypropylene.

From the characterization requirements (a) to (c) as listed above, it can be confirmed that the stereoregular polypropylenes of the present invention do not contain substantially chains of different bond and racemic bond and demonstrate exceedingly high isotactic characteristics consisting of much highly controlled meso bond chains.

Further, the term "terminal double bond" as used herein refers to the proportion of the double bond present at the end of polypropylene molecule determined by ^{13}C NMR spectra in accordance with the method suggested by T. Hayashi et al in POLYMER Vol. 30, 1714-1722, 1989.

The requirement (d) for characterizing the stereoregular polypropylene of the present invention is the absence of the terminal double bond. If a double bond is present at the end of polypropylene molecules, a chemical stability of polypropylene may be impaired due to the participation of the double bond in the reaction depending on the use conditions, which will result in no occurrence of inherent characteristics of polypropylene.

Of the requirements for characterization of the present stereoregular polypropylene, (e) a weight average molecular weight (Mw) and (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) are calculated from the results determined by a gel permeation chromatography (GPC) in the following manner.

They were measured at 135°C using an o-dichlorobenzene solution with 0.05% by weight of a polymer concentration and a mixed polystyrene gel column, e.g. PSK gel GMH6-HT available from Toso K.K. in Japan. As a measuring device, GPC-150C manufactured by Waters Co. Ltd. is used for instance.

The requirements for characterization of the present stereoregular polypropylene, (e) a weight average molecular weight (Mw) is 50,000 to 1,000,000, preferably 100,000 to 1,000,000.

The requirements for characterization of the present stereoregular polypropylene, (f) a ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is 1.5 to 3.8, preferably 1.5 to 3.5. The ratio (Mw/Mn) of the weight average molecular weight (Mw) to a number average molecular weight (Mn) is a measure of a molecular weight distribution. Larger ratio (Mw/Mn) means wider molecular weight distribution, whereas smaller ratio means narrower molecular weight distribution.

The requirements for characterization of the present stereoregular polypropylenes are six requirements as mentioned above. A melting point of the present polypropylene shows 160 to 168°C due to these structural feature, in particular, the above requirements (a) to (c) and 161 to 168°C, further 162 to 168°C depending on the structural condition.

The melting point as referred to herein is a temperature showing a peak on melting which was determined by heating polypropylene from room temperature to 230°C at a rate of 30°C/min, keeping it at the same temperature for 10 minutes, followed by cooling down to -20°C at a rate of -20°C/min, keeping it at the same temperature for 10 minutes and heating again it at a rate of 20°C/min, using a DSC 7 type differential scanning calorimeter manufactured by Perkin Elmer Co.

If the stereoregular polypropylene of the present invention satisfies the above-mentioned requirements for characterization, the processes for the production thereof are not limited. For instance, the process using a specified metallocene catalyst will be explained below.

The metallocene catalyst used in this process is the catalyst using metallocene as a transition metal compound and comprising combination thereof with aluminoxane.

Those which can be used as the above metallocene can include chiral transition metal compounds represented by the formula of $\text{Q}(\text{C}_5\text{H}_{4-m}\text{R}^1_m)(\text{C}_5\text{H}_{4-n}\text{R}^2_n)\text{MXY}$ wherein $(\text{C}_5\text{H}_{4-m}\text{R}^1_m)$ and $(\text{C}_5\text{H}_{4-n}\text{R}^2_n)$ represent a substituted cyclopentadienyl group; m and n are an integer of 1 to 3; R^1 and R^2 may be the same or different and each represents a hydrocarbyl radical of 1 to 20 carbons, a silicone-containing hydrocarbyl radical or a hydrocarbyl radical which is joined with two carbon atoms on the cyclopentadienyl ring to form one or more hydrocarbon ring which may be substituted by a hydrocarbon; Q is a group capable of crosslinking $(\text{C}_5\text{H}_{4-m}\text{R}^1_m)$ and $(\text{C}_5\text{H}_{4-n}\text{R}^2_n)$ and represents a divalent, hydrocarbyl radical, an unsubstituted silylene group or a hydrocarbyl-substituted silylene group; M represents a transition metal selected from the group consisting of titanium, zirconium and hafnium; X and Y may be the same or different and each represents a hydrogen, a halogen or a hydrocarbyl radical.

More preferably, the chiral transition metal compounds are used wherein R^1 and R^2 may be the same or different and each represents an alkyl group of 1 to 20 carbons, Q is a dialkyl silylene group, M represents a transition metal of zirconium or hafnium, and X and Y may be the same or different and each represents a halogen or a hydrocarbyl radical.

Examples of such metallocenes include:

rac-Dimethylsilylene bis(2-methyl-4,5,6,7-tetrahydroindenyl)zirconium dichloride,
rac-Dimethylsilylene bis(2-methyl-4,5,6,7-tetrahydroindenyl)zirconium dimethyl,
rac-Ethylene bis(2-methyl-4,5,6,7-tetrahydroindenyl)hafnium dichloride,

rac-Dimethylsilylene bis(2-methyl-4-phenylindenyl)zirconium dichloride,
 rac-Dimethylsilylene bis(2-methyl-4-phenylindenyl)zirconium dimethyl,
 rac-Dimethylsilylene bis(2-methyl-4-phenylindenyl)hafnium dichloride,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)titanium dichloride,
 5 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)zirconium dichloride,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)zirconium dichloride,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)zirconium dimethyl,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)hafnium dichloride,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)hafnium dimethyl,
 10 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)titanium dichloride,
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)zirconium dichloride,
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)zirconium dimethyl,
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)hafnium dichloride, and
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)hafnium dimethyl.

Of these metallocenes, especially preferred are the following compounds:

Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)zirconium dichloride,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)zirconium dimethyl,
 Dimethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)hafnium dichloride,
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)zirconium dichloride,
 20 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)zirconium dimethyl,
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)hafnium dichloride, and
 Dimethylsilylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)hafnium dimethyl.

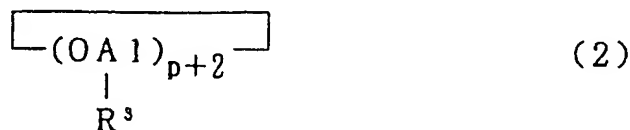
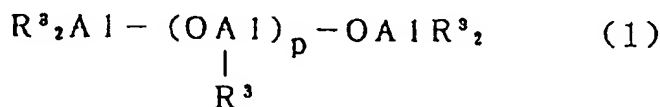
In the synthesis of these chiral metallocenes, a metallocene of meso form in a non-chiral structure may be formed as a by-product. In the practical use, however, all are not required to be chiral metallocenes and the meso form may be mixed. When a mixture with the meso form is used, there may be the case where atactic polypropylene polymerized from the meso form is required to remove by known process such as solvent extraction or the like so that the resulting polypropylene may meet the essential requirements of the present invention depending on the proportion of the meso form mixed and the polymerization activity of propylene.

Those chiral metallocenes can be formed in combination with aluminoxane into a catalyst, but can be supported on a finely divided carrier. The carrier is an inorganic or organic compound. The finely divided solid in a granular or spherical form having a particle diameter of 5 to 300 μm , preferably 10 to 200 μm is used.

The inorganic compounds used for the carrier include SiO_2 , Al_2O_3 , MgO , TiO_2 , ZnO or the mixtures thereof, e.g. $\text{SiO}_2\text{-Al}_2\text{O}_3$, $\text{SiO}_2\text{-MgO}$, $\text{SiO}_2\text{-TiO}_2$, $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-MgO}$. Of these compounds, those comprising SiO_2 or Al_2O_3 as a main component are preferred.

The organic compounds used for the carrier include polymers or copolymers of α -olefins of 2-12 carbons such as ethylene, propylene, 1-butene, 4-methyl-1-pentene, and polymer or copolymer of styrene.

Aluminoxane as one of the catalyst components which can be used in combination with metallocene in the process for the production of high stereoregular polypropylenes of the present invention is an organic aluminum compound represented by the following formula (1) or (2).



wherein R^3 represents a hydrocarbyl radical of 1 to 6 carbons, preferably 1 to 4 carbons, which includes an alkyl group such as methyl, ethyl, propyl, butyl, isobutyl, pentyl, hexyl; an alkenyl group such as allyl, 2-methylallyl, propenyl, isopropenyl, 2-methyl-1-propenyl, butenyl; a cycloalkyl group such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl; and an aryl group. Of these, the alkyl group is especially preferred and each R^3 may be identical or different. p is an integer of 4 to 30, preferably 6 to 30, especially preferably 8 to 30.

These aluminoxanes can be used singly or in combination of two or more. Also, they can be used in admixture with an alkyl aluminum compound such as trimethyl aluminum, triethyl aluminum, tri-isopropyl aluminum, tri-isobutyl alumi-

num, dimethyl aluminum chloride.

The above-mentioned aluminoxanes can be prepared under various known conditions. More specifically, the following methods can be illustrated:

- (1) a method of reacting a trialkyl aluminum directly with water in the presence of an organic solvent such as toluene, ether;
- (2) a method of reacting an trialkyl aluminum with salts containing water of crystallization such as copper sulfate hydrate, aluminum sulfate hydrate;
- (3) a method of reacting an trialkyl aluminum with water impregnated in silica gel or the like;
- (4) a method of reacting a mixture of trimethyl aluminum and tri-isobutyl aluminum directly with water in the presence of an organic solvent;
- (5) a method of reacting a mixture of trimethyl aluminum and tri-isobutyl aluminum with salts containing water of crystallization such as copper sulfate hydrate, aluminum sulfate hydrate; and
- (6) a method of reacting tri-isobutyl aluminum with water impregnated in silica gel, followed by reacting with trimethyl aluminum.

The catalyst which can be used in the process for the production of high stereoregular polypropylenes according to the present invention is the catalyst consisting of combination of metallocene and aluminoxane. The proportion of each catalyst component used is in such a range that an aluminum atom in aluminoxane is from 10 to 100,000 mol, preferably 50 to 50,000 mol, especially preferably 100 to 30,000 mol per mol of a transition metal atom in metallocene.

The polypropylenes of the present invention are produced by the polymerization of propylene using the thus combined catalyst in the presence of hydrogen. As a process for the polymerization of propylene can be used known polymerization processes of propylene. Those processes include a slurry polymerization wherein propylene is polymerized in an inert solvent including an aliphatic hydrocarbon such as butane, pentane, hexane, heptane, isooctane; an alicyclic hydrocarbon such as cyclopentane, cyclohexane, methylcyclohexane; an aromatic hydrocarbon such as toluene, xylene, ethylbenzene; and gasoline fraction and hydrogenated diesel oil; a bulk polymerization wherein propylene itself is used as a solvent; and a gas phase polymerization wherein propylene is polymerized in a gas phase.

As the catalyst in the polymerization of propylene, a mixture obtained by previously mixing both components of metallocene and aluminoxane in the inert solvent may be fed to a polymerization reaction system, or metallocene and aluminoxane may be separately fed to the reaction system. Prior to the polymerization of propylene, the catalyst consisting of combination of metallocene and aluminoxane may be pre-activated by the polymerization reaction of said catalyst with small amounts of α -olefins, more specifically about 1 g to about 10 kg of α -olefins per mole of the transition metal in metallocene, and subsequently the polymerization of propylene can be carried out. This procedure is effective in obtaining a final polypropylene in good particular form, which is included within the scope of the present invention.

As α -olefins which can be used in the pre-activation of the catalysts, there are preferably used those of 2 to 12 carbons which include ethylene, propylene, butene, pentene, hexene, octene, 4-methyl-1-pentene or the like. In particular, ethylene, propylene and 4-methyl-1-pentene are preferably used.

The thus prepared catalysts or the pre-activated catalysts are used in the polymerization of propylene according to the polymerization processes as mentioned above. As the polymerization conditions can be employed similar conditions to those in the polymerization of propylene according to known conventional Ziegler catalysts. More specifically, the polymerization of propylene is performed at the polymerization temperature ranging from -50 to 150°C, preferably -10 to 100°C and the polymerization pressure ranging from an atmospheric pressure to 7 MPa, preferably 0.2 to 5 MPa in the presence of hydrogen, usually for about one minute to about 20 hours. The suitable amount of hydrogen is 0.01 kPa to 5 MPa, preferably 0.1 kPa to 3 MPa in terms of a partial pressure of hydrogen at the gas phase portion in the polymerization reactor.

After completion of the polymerization of propylene, if necessary, known after-treatments may be carried out, including deactivation of the catalyst, removal of the catalyst residue, drying of the product or the like. The high stereoregular polypropylenes of the present invention are produced through such after-treatments, but must have six requirements for characterization as listed hereinabove. If such requirements are not satisfied, the object of the present invention cannot be achieved.

Thus, the polymerization of propylene using the above metallocene under the above-mentioned polymerization conditions cannot always produce the desired high stereoregular polypropylenes of the present invention. Depending on the kind of metallocene, choice of an optimum polymerization condition is required. For the optimum condition, relatively lower polymerization temperature is often selected from the range of the polymerization conditions as mentioned above.

High stereoregular polypropylenes of the present invention as produced in this manner have narrow molecular weight distribution and higher melting point. The present high stereoregular polypropylenes can be served as a molding compound by compounding with various additives such as antioxidants, ultraviolet absorbing agents, antistatic agents, nucleating agents, lubricants, flame retardants, antiblocking agents, colorants, inorganic or organic fillers or the like, if



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 96 10 8226

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
Y,D	DATABASE WPI Derwent Publications Ltd., London, GB; AN 91-062694 XP002013308 & JP-A-03 012 407 (CHISSO) , 21 January 1991 * abstract; example 3 *	1-11	C08F10/06
Y	US-A-4 962 262 (WINTER ANDREAS ET AL) 9 October 1990 * column 1, line 60 - column 2, line 24 * * column 5, line 34 - line 41 *	1-11	
X	EP-A-0 485 822 (HOECHST AG) 20 May 1992 * page 9, line 13 - line 15 * * examples 22,32 *	1-11	
X	EP-A-0 629 632 (MITSUI PETROCHEMICAL IND) 21 December 1994 * page 20, line 45 - page 21, line 25 *	1-11	
A	JOURNAL OF APPLIED POLYMER SCIENCE, vol. 52, 1993, NEW YORK, pages 159-172, XP000449415 A.MIZUNO ET AL: "structural features of isotactic polypropylene obtained by ethylenebis(a-indenyl)hafniumdichloride/methylaluminoxane catalyst system" * tables II,III *	1-11	TECHNICAL FIELDS SEARCHED (Int.Cl.6) C08F
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 13 September 1996	Examiner Schmidt, H
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure F : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

EPO FORM 1503 03/92 (P04CH)

FIG. 1

